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Dedicated to B.I. Buzykin on His 80th Anniversary

Azomethines Based on Pyridoxal-Derived Aromatic Aldehydes

L. K. Kibardina[a]*, A. V. Trifonov[a,c], Yu. I. Ivanova[c], M. A. Pudovik[a], E. M. Pudovik[b], and A. R. Burilov[a,c]

[a] *Arbuzov Institute of Organic and Physical Chemistry, Kazan Scientific Center, Russian Academy of Sciences, ul. Arbuzova 8, Kazan, Tatarstan, 420088 Russia*
**e-mail: kibardina@iopc.ru*

[b] *Kazan (Volga Region) Federal University, Kazan, Tatarstan, Russia*

[c] *Kazan National Research Technological University, Kazan, Tatarstan, Russia*

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Abstract—The reaction of pyridoxal with 2,4-dihydroxybenzaldehyde in a hydrochloric acid solution gave 1-(5-formyl-2,4-dihydroxyphenyl)-7-hydroxy-6-methyl-1,3-dihydrofuro[3,4-*c*]pyridin-5-ium chloride. Treatment of the latter with sodium hydride in ethanol afforded a free aldehyde in 85% yield. A series of azomethines and imidazolidines was obtained by reacting the obtained aldehyde with various amines and diamines.

Keywords: pyridoxal, 2,4-dihydroxybenzaldehyde, polycyclic aromatic aldehyde, amines, diamines, azomethines, imidazolidines

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Functionalization of biologically active compounds, vitamins among them, is an effective approach to substances with useful properties. In this regard, the cyclic derivatives of pyridoxal (one of the components of vitamin B_6) like furopyridines containing an aryl substituent in the furan ring are of undoubted interest. Some of specimens of this class of compounds have a variety of pharmacological activities [1–3]. 2,4-Dihydroxybenzaldehyde-derived compounds with a double C=N bond are widely used for obtaining polymers, liquid crystals, and other materials with practically useful properties [4–6]. The influence of the solvent nature on the tautomeric processes in these objects has been studied in [7, 8]. It should also be noted that azomethines, including 2,4-dihydroxybenzaldehyde derivatives, exhibit a wide range of biological activity: fungicidal, antibacterial, antimalarial, anti-inflammatory, antiviral, antipyretic [9], anticarcinogenic [10].

Previously we have developed a new method for the synthesis of 1-arylfuropyridines by condensation of pyridoxal with a number of phenols and polyphenols in an alcoholic medium in the presence of hydrochloric acid [11]. Resorcinol, methylresorcinol, pyrocatechol, pyrogallol, resorcinic acid, phenol have been used as the hydroxyl-containing aromatic compounds. Taking into account these data, we tried to obtain a polycyclic compound bearing pyridoxal fragment and a reactive formyl group. Thus, 2,4-dihydroxybenzaldehyde **1** was involved into the reaction with pyridoxal. However, it was not possible to isolate the target product when using the reaction conditions reported earlier. Therefore, we changed the procedure and carried out the reaction in a solution of hydrochloric acid. Under these conditions, aldehyde **1** reacts with pyridoxal hydrochloride **2** to form two products: 1-(5-formyl-2,4-dihydroxyphenyl)-7-hydroxy-6-methyl-1,3-dihydrofuro[3,4-*c*]pyridin-5-ium chloride **3** and 1-(3-formyl-2,6-dihydroxyphenyl)-7-hydroxy-6-methyl-1,3-dihydrofuro[3,4-*c*]pyridin-5-ium chloride **4** in a 1 : 1 ratio, i. e., the reaction affects both the third and fifth positions relative to the formyl group of the aromatic ring of compound **1** (Scheme 1).

The process was completed within 2 h; the yield of aldehyde **3** crystallizing from the reaction mixture was 50.3%. The isolation of the second product **4** involved certain difficulties. Initially, it remains in the acid solution and crystallizes from it slowly when the acid